Mechanisms for Stable Sonoluminescence

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(February 5, 2008)

A gas bubble trapped in water by an oscillating acoustic field is expected to either shrink or grow on a diffusive timescale, depending on the forcing strength and the bubble size. At high ambient gas concentration this has long been observed in experiments. However, recent sonoluminescence experiments show that in certain circumstances when the ambient gas concentration is low the bubble can be stable for days. This paper presents mechanisms leading to stability which predict parameter dependences in agreement with the sonoluminescence experiments.

Recent experiments on sonoluminescence (SL) [1–7] allow detailed studies of the dynamics of a bubble levitated in a periodically modulated acoustic field. Besides the light emission itself, one of the greatest mysteries is how the bubble can exist in a stable state for many billions of cycles. Measurements of the time between successive light flashes show that the total mass of the bubble remains constant to high accuracy [1,2,7]. This result contradicts classical notions about the dynamics of periodically forced bubbles: An unforced bubble of ambient radius R_0 dissolves over a diffusive timescale, $\tau \sim \frac{\rho_0 R_0^2}{D(c_0 - c_\infty)}$ [8], where ρ_0 is the ambient gas density in the bubble, D is the diffusion constant of the gas in the liquid, c_0 is the saturated concentration of the gas in the liquid, and c_∞ is the concentration of gas in the liquid far from the bubble. A strongly forced bubble grows by rectified diffusion, as first discovered by Blake [9,11]. This is because when the bubble radius is large, the gas pressure in the bubble is small, resulting in a strong mass flux into the bubble. Conversely, when the bubble radius is small there is a strong mass outflux. Since the diffusive time scale is much larger than the very short time the bubble spends at small radii, gas cannot escape from the bubble during the compression phase and will be recollected during expansion, so that the net effect is bubble growth. At a special value of the ambient radius R_0^* rectified diffusion and normal diffusion exactly balance. However the above arguments suggest that this equilibrium point is *unstable*; if the ambient radius is infinitesimally different from R_0^* the bubble is pushed away from equilibrium.

The classic papers on rectified diffusion (see e.g. Eller and Crum [10–12]) verified the qualitative picture described above when $c_\infty/c_0 \approx 1$. Anomalies between theory and experiment do however exist: of special note is Eller's [10] observation of a stable oscillating bubble persisting over long periods.

There are two controlled parameters in the SL experiments: the forcing pressure P_a and the gas concentration c_∞. The key to the discovery of *stable* single bubble SL (whose existence completely contradicts the above scenario) by Gaitan et al. [1] was that (i) $c_\infty/c_0 \ll 1$, and (ii) P_a must lie between a lower critical pressure ≈ 1.1atm and an upper critical pressure ≈ 1.3atm. When P_a is within this window of stability, the ambient radius remains constant for billions of cycles, as evidenced by the constant phase ϕ of the light emission relative to the oscillatory forcing. Barber and Putterman [2] showed that the "jiggle" in the phase differs by less than 50 picoseconds from cycle to cycle. Outside the window of stability, ϕ (and hence R_0) varies on a diffusive timescale [5]. The phase grows (implying growth of the ambient radius), until the bubble becomes parametrically unstable [13] and microbubbles pinch off. Experiments [5] show that ϕ can oscillate indefinitely on a diffusive timescale via this mechanism (diffusive growth followed by pinching off a microbubble).

The dependence of stable SL on the gas concentration c_∞ is demonstrated by the UCLA experiments on pure argon bubbles [4,5]. For c_∞/c_0 between approximately 0.06 and 0.25, ϕ oscillates on a diffusive timescale as described above. At lower argon concentration $c_\infty/c_0 = 0.004$ however the phase becomes perfectly stable [5], indicating a stable equilibrium.

The goal of this paper is to suggest mechanisms leading to stabilization: When c_∞/c_0 is decreased at forcing pressures $P_a > 1.1$atm, the classical unstable equilbrium point R_0^* undergoes an inverse pitchfork bifurcations and actually *stabilizes.* At even higher forcing pressures, there can be *several* stable fixed points, although far from the equilibrium point small bubbles shrink and large bubbles grow. This mechanism is sufficient to explain the stable equilibria in the SL experiments. However, high pressures and temperatures within the bubble cause non-diffusive effects [7] which can also stabilize the bubble. Both mechanisms predict parameter dependences consistent with SL experiments. We suggest that the discretization of the ambient radius predicted by the diffusive mechanism provides a clear experimental signature

as to which effect is primarily responsible for stable SL.

We first set up a formalism for studying the stability of the equilibrium point, following Fyrillas and Szeri [14] and Löfstedt et al. [7]. Let $c(r,t)$ denote the concentration of gas dissolved in the liquid a distance r from the center of the bubble. For $r > R(t)$, where $R(t)$ is the radius of the bubble, c satisfies a convection diffusion equation

$$\partial_t c + \frac{R^2 \dot{R}}{r^2}\partial_r c = D\nabla^2 c. \tag{1}$$

The boundary conditions are given by Henry's law $c(R,t) = c_0 P(R,t)/P_0$ and by $c(\infty,t) = c_\infty$. The concentration gradient at the boundary gives the mass loss/gain of the bubble $\dot{M} = 4\pi R^2 D \partial_r c|_{R(t)}$.

These equations determine the growth of the bubble as a function of time. There are two crucial observations: First, Eller noted that changing coordinates to $h = \frac{r^3 - R^3}{3}$ and $\tau = \int^t R^4 dt$ transfers equation (1) to the simpler form

$$\partial_\tau c = D\partial_h\left(\left(1 + \frac{3h}{R^3}\right)^{4/3}\partial_h c\right) = 0. \tag{2}$$

For the following it is convenient to define the τ-average of a function $f(t)$ by $\langle f(t)\rangle_\tau = \int f(t)R(t)^4 dt / \int R(t)^4 dt$.

The second observation [14,7] is that the bubble radius changes over a much faster timescale than the ambient radius. By averaging equation (2) over the fast time scale, $\partial_h c(\tau)$ can be computed on the slower diffusive timescale. Then the dynamics of the ambient radius is given by

$$R_0^2 \frac{dR_0}{d\tau} = D\frac{c_\infty - \langle c\rangle_\tau}{\int_0^\infty \frac{dh}{<1+\frac{3h}{R^3}>_\tau}}. \tag{3}$$

Equilibrium points satisfy

$$\frac{\langle p\rangle_\tau}{P_0} = \frac{c_\infty}{c_0}. \tag{4}$$

The equilibrium is stable if the quantity $\beta = \frac{d\langle p\rangle_\tau}{dR_0}$ is positive.

Now we proceed to analyze this model. We calculate numerically $\langle p\rangle_\tau$ as a function of R_0, for different driving pressures, by τ averaging solutions $R(t)$ of the Rayleigh – Plesset (RP) equation. The RP equation [6,15,16] governs the dynamics of an acoustically forced bubble, and is given by

$$R\ddot{R} + \frac{3}{2}\dot{R}^2 = \frac{1}{\rho_w}\left(p(R,t) - P(t) - P_0\right) + \frac{R}{\rho_w c_w}\frac{d}{dt}\left(p(R,t) - P(t)\right) - 4\nu\frac{\dot{R}}{R} - \frac{2\sigma}{\rho_w R}. \tag{5}$$

We use parameters corresponding to [17,3,6] an air bubble in water: the surface tension of the air-water interface is $\sigma = 0.073kg/s^2$, while water has viscosity $\nu = 10^{-6}m^2/s$, density $\rho_w = 1000kg/m^3$, and speed of sound $c_w = 1481m/s$. The acoustic field is driven via $P(t) = P_a\cos(\omega t)$ with $\omega/2\pi = 26.4kHz$ and external pressure $P_0 = 1$atm. The pressure inside the bubble varies adiabatically like $p(R) \sim (R^3 - a^3)^{-1.4}$. Here $a = R_0/8.73$ is the hard core van der Waals radius.

Figure 1 shows $\langle p\rangle_\tau/P_0$ for several values of P_a. For small P_a , $\langle p\rangle_\tau$ monotonically decays with R_0, signaling a diffusively *unstable* equilibrium. For example, when $c_\infty/c_0 = 1$ with a forcing amplitude of $P_a = 0.8$ the unstable equilibrium occurs at $R_0 \approx 5\mu m$. Note that for large R_0 the bubble may become unstable with respect to shape oscillations [13].

At large P_a however $\langle p\rangle_\tau$ develops oscillations as a function of R_0, so for a range of c_∞/c_0 there are several stable equilibrium points. These stable equilibria only occur at low c_∞/c_0, immediately suggesting a reason why diffusively stable SL only occurs under these conditions. As an example, see the inset of figure 1: when $c_\infty/c_0 = 10^{-2}$ and $P_a = 1.25$atm, there are stable equilibria (denoted by small dots in the figure) at $R_0 = 6.5, 6.8, 7.1, 7.5, 8.0$ and 8.5μm. To further verify the existence of multiple stable equilibria, we have solved the full equations (1) and (5) numerically with a standard finite difference scheme [18]. Figure 2 shows the ambient radius as a function of time for two different initial conditions with $P_a = 1.25$atm. In each case, the ambient radius saturates towards a constant (7.1μm and 8.4μm) at long times.

We now outline in detail the predictions of these calculations for the SL experiments. A standard experimental protocol [1,3] is to slowly increase the driving pressure P_a. The initial ambient radius depends on the preparation of the bubble. At low pressures, if the bubble size is below the diffusive equilibrium curve sketched in figure [**?**], the bubble shrinks. As the forcing pressure is increased, there is a critical pressure where the bubble size becomes greater than R_0^*; the calculations for $c_\infty/c_0 = 0.25$ indicate this occurs near 1atm, in accord with experiments [3]. Above this forcing pressure, the bubble grows by rectified diffusion. When the ambient radius becomes too large the bubble is parametrically unstable; in experiments the bubble decreases its radius by pinching off a microbubble (indicated by the downward arrows in figure 2). As the forcing pressure is further increased, the bubble size tracks the parametric instability line [13]. However, at the pressure $P_a \approx 1.1$atm stable equilibrium points appear. A sketch of this inverted pitchfork bifurcation in the $R_0 - P_a$ phase diagram is shown in figure 3. If the bubble is attracted to one of the stable points the radius stabilizes, causing a discontinuous jump in the ambient radius, as observed by Barber *et. al.* [3] for air bubbles. This analysis predicts a similar jump also for argon bubbles. The stable state persists for $1.1 < P_a < 1.3$; Above $P_a = 1.3$ the equilibrium point destabilizes, and the bubble must return

to diffusive growth followed by microbubble pinching to survive. For even larger P_a the bubble becomes unstable with respect to shape oscillations [13]. This entire scenario suggested by the classical equations of diffusive dynamics is in good agreement with the findings from the SL experiments.

The stable equilibrium points are ultimately due to oscillations in $\langle p \rangle_\tau$ as a function of R_0, which arise from resonances in the Rayleigh-Plesset equation. Oscillations even occur in the maximum radius as a function of R_0, so that in some situations adding more gas to a bubble decreases its maximum size. A comparison with the Mathieu equation is instructive: If the eigenfrequency (in the RP equation this depends on R_0) is an integer or half integer fraction of the forcing frequency, the amplitude of the oscillations is anomolously large.

Our predictions are in qualitative agreement with the experiment; precise quantitative agreement requires accounting for several neglected effects. These include realistic heat transfer [19,6] and equations of state for the gas [20], as well as shocks within the bubble [21,20], which cause the light emission itself.

Perhaps more importantly, the high pressures and temperatures within the bubble cause complications which influence the position and stability of the equilibria. For example, the RP equation predicts that the minimum pressure inside the bubble can become as low as 10^{-3}atm. However, the presure cannot fall below the equilibrium vapor pressure of water around 10^{-2}atm. Thus, the RP equation can drastically underestimate the pressure inside the bubble near its maximum radius. Calculations where the pressure inside the bubble is given by $max(0.05 \text{ atm}, P(R(t)))$ show that this shifts the position of the equilibrium point to larger R_0, without affecting the stability.

The increase of the mass diffusion constant with increasing temperature and pressure affects both the stability and the location of the equilibrium point: When the pressure and the temperature inside the bubble is high, the diffusion constant near the bubble wall is larger than the diffusion constant in the bulk liquid. This results in nondiffusive mass ejection when the bubble is small [7].

We study this increase in the interfacial diffusion constant with an extremely simple model: Whenever the pressure inside the bubble exceeds a critical pressure $p_{thres}(R)$ we discontinuously increase the diffusion constant near the bubble wall by a factor f_{thres}. The diffusion constant in the bulk liquid remains constant, since high pressures and temperatures are localized near the bubble wall.

Numerical simulations of the full equations [18] with and without this effect demonstrate that the unstable equilibrium point can be stabilized by this effect. The position of the equilibrium point is in general shifted to larger radii. Although it is difficult to determine the precise parameter dependence of this mechanism without a more accurate model for the diffusion constant, it clearly only operates at the high pressures where sonoluminescence occurs.

To summarize, we have presented two different mechanisms leading to stabilization of the ambient bubble radius. Contrary to classical intuition, the diffusive dynamics themselves have *multiple* stable equilibrium points in the SL regime. The dependence of the mass diffusion constant on temperature and pressure also leads to stabilization. Which effect dominates the SL experiments? We suggest that this issue can be settled experimentally by determining whether the ambient radius takes on only a discrete set of values, as predicted by the purely diffusive mechanism.

Acknowledgements: We thank S. Grossmann, S. Hilgenfeldt, and L. Kadanoff for helpful discussions. This work has been supported by the DOE, the MRSEC Program of the National Science Foundation at the University of Chicago, and by the DFG through the SFB 185.
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FIG. 1. $\langle p\rangle_\tau/P_0$ as a function of R_0 (in μm) for P_a = 0.8atm, 0.9atm,1.atm,1.1atm, 1.2atm and 1.25atm, top to bottom. Equilibrium corresponds to $\langle p\rangle_\tau/P_0 = c_\infty/c_0$. The equilibrium is diffusively stable if the slope $\beta = d\langle p\rangle_\tau/dR_0$ is positive. *Inset:* An enlargement of P_a = 1.25atm. The straight line corresponds to $c_\infty/c_0 = 10^{-2}$. The intersection of the straight line with the curve correspond to equilibrium points. When $\beta > 0$ (the solid dots in the figure) the equilibrium is stable.

FIG. 2. The ambient radius $R_0(t)$ as a function of time, for P_a = 1.25atm and $c_\infty/c_0 = 0.01$, with two different initial ambient radii. Each bubble approaches a *different* equilibrium ambient radius, demonstrating that more than one stable equilibrium may exist.

FIG. 3. Sketch of the stability diagram in the $R_0 - P_a$ phase space. Upon increasing the forcing P_a, a window of diffusive stability develops through an inverse pitchfork bifurcation. Further bifurcations can occur. At larger P_a the stability window closes. Stable and unstable branches are marked by s and u. The upper curve shows the parametric instability line. The long arrows sketch growth by rectified diffusion followed by micro bubble pinchoff.